

XPM nonlinearities:generation of cross field noise correlations due to a novel symmetry

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Many symmetries of nonlinear susceptibilities[1],especially in crystals,are well known.We have found a novel symmetry for XPM(Cross Phase Modulation) systems .We prove that this symmetry is a necessary condition for a single multiphoton process to be visible in more than one field.We have found, two EIT(Electromagnetically Induced Transparency) systems that show cases of this symmetry.The above process has the possibility of leading to strong sources of temporally entangled photons of different frequency. For cases that do not show this symmetry,more than one "species" of multiphoton processes can take place and each one is visible in one field only.However,an asymmetric case might be seen as a sum of the symmetric and asymmetric part. Whether or not the symmetry is a sufficient condition for a single multiphoton process to be visible in more than one field, we predict cross field noise correlations for the scattered fields.Cross field noise correlations have recently been seen in a different situation that can provide interesting variation to our study,namely the generated fields in FWM(Four Wave Mixing) [3],[4],[2]. We postulate or conjecture the existence of two types of asymmetric multiphoton processes. One of systems we use for this purpose generates a $\chi^{(9)}$ absorption resonance that is observable on the same trace that shows linear features, in an experiment described in a previous work[18] but the order of nonlinearity newly interpreted using this work.This high order nonlinearity should be very sensitive to cross field correlations.This can be used to test our conjecture of asymmetric processes. We also come across an important $\chi^{(5)}$ absorption,comparable to linear absorption, that is anomalous.The anomaly is that the Imaginary part of this XPM nonlinearity can cause squeezing.We give a qualitative argument which shows that we don't expect the Imaginary part of the XPM nonlinearities we usually come across, to cause squeezing.This promising source of squeezing, is qualitatively different from the other sources hitherto known.

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I. INTRODUCTION

Symmetries are sought after as they make the analysis of a system easier.There is more than one symmetry of nonlinear susceptibilities seen, especially in crystals, like the Kleinman's symmetry.The symmetry we have found is a necessary condition for a single multiphoton process to be visible in more than one field in XPM[15] systems. The advent of lasers in 1960's produced a lot of research in the field of multiphoton processes, that absorb multiple photons from a single beam, or SPM nonlinearities.Topics like stepwise nonlinear processes[5] as opposed to the simultaneous process that this work is about,field correlation sensitivity [6],squeezing[7] have been studied. XPM multiphoton processes became easily accessible after 1990 with the discovery[9] of enhancement in the presence of EIT, of these nonlinearities.The advantage of this enhancement is great as it can, in some cases, make them comparable in strength to linear processes at much lower intensities compared to that at which SPM processes are observable. XPM multiphoton processes are qualitatively different because of effects that arise from the absorption(emission) of photons forming the chain, belonging to different fields.This changes properties like squeezing and field correlation sensitivity.In fact, the difference raises a basic question,relevant to many recent studies[3],[4],[2], whether the nonlinear susceptibilities faced by different fields, due to a single "species" of multiphoton process, bear any relation to each other as the quanta of light absorbed by them is related? We address it in this work. We show in sectionV the *generation of cross field correlation* in the different fields as the result of the symmetry that the nonlinear susceptibilities, due to a single "species" of multiphoton process, faced by different fields are the same. We have found, from our calculations, cases which show this symmetry in the two atom-laser systems shown in the figures [1], [2].

The XPM nonlinearity in the probe beam in the system1 described in figure [1] has been

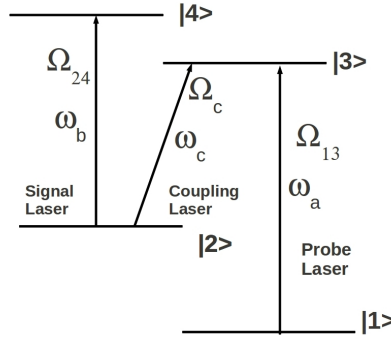


FIG. 1: This level scheme represents system1 and has been considered theoretically by Schmidt et.al.[8].This can represent the D2 line of Rb^{87} system given, for example, in [16].

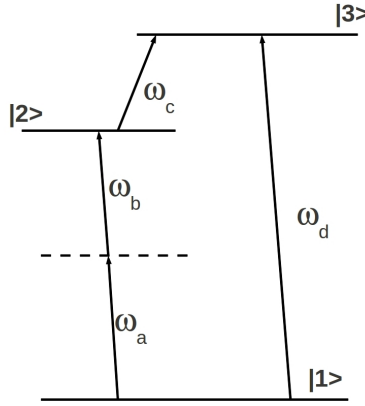


FIG. 2: This level scheme is system2 and has been considered theoretically by Harris et. al.[9]

observed by Kang and Zhu [16]. Three out of roughly a dozen of the XPM nonlinearities considered in this work, in systems given by figures [1], [2] have been long known and have been considered before theoretically by Schmidt and Imamoglu [8] and Harris et. al. [9] respectively. Harris et.al., without giving any reasons, have brought to notice first one of the symmetries though, from the analysis we have made, we agree only partially about it's actually being a symmetry. In fact, this example of "partial symmetry" turns out to be useful as it shows how an asymmetric case can be seen as the sum of symmetric and asymmetric parts.

The hitherto well known EIT enhanced XPM nonlinearities are of the type where the polarization is proportional to $\chi^{(x+y+z)} E_1^x E_2^y E_3^z$ where $x=1$ or 0 and y, z can be greater than 1 , where E_1 is the field that is being observed. $x=0$ or 1 , implies that these well known nonlinearities cannot cause light squeezing by using the Imaginary part. There is however a system, we have found, for which $x=5, y=z=0$. It should be possible to see light squeezing in it, using the Imaginary part of the nonlinearity.

To see that the Imaginary part of every XPM nonlinear susceptibility starting from $\chi(2)$ does not cause squeezing, consider a nonlinearity $\chi^{(2)} E^2$ where E is the field in which it is observed. The intensity of the field, recorded in a detector will not scale down linearly because

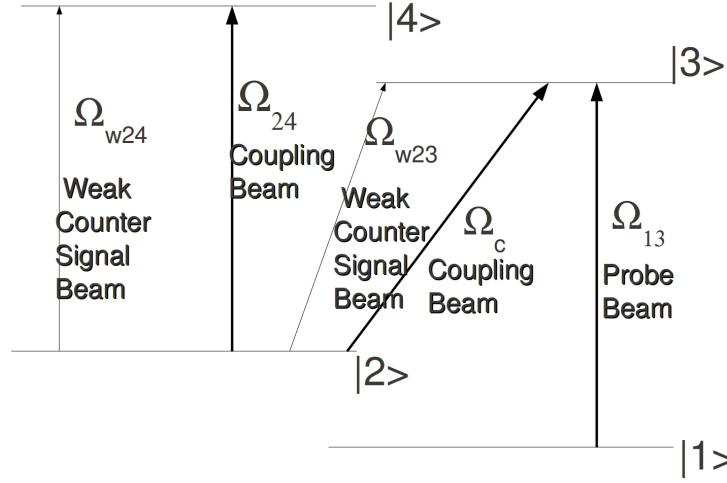


FIG. 3: This level scheme represents system3 and is a modified scheme based on an experiment done before described in [18]. The weak signal beam is considered as a perturbation.

of the nonlinear medium. It scales down slightly more where there is a positive fluctuation in intensity above the mean and scales down less where there is a negative fluctuation in the intensity as is clear from the fact that $11^2 - 10^2 = 21$ while $10^2 - 9^2 = 19$. This should be a necessary condition for the Imaginary part of a susceptibility to cause squeezing. Now consider a nonlinearity $\chi^{(3)} E_1 E_2^2$ which is being observed in the field E_1 . Since the shot noise pattern of field E_2 is different from that of E_1 the points that are scaled down more and are scaled less do not necessarily correspond to a positive and negative fluctuation respectively of E_1 . Hence it cannot lead to squeezing.

Whatever light squeezing, in an atomic medium, that is known to be possible till now relies on an origin qualitatively different from that of the $x=5$ nonlinearity. Light squeezing due to atomic Kerr nonlinearity, for example, relies mainly on $\chi^{(3)}$ nonlinearities due to saturation and power broadening effects in resonant media that is put in a cavity [10], [11]. It is also different from SPM nonlinearities in that it has large values at low light levels. However, a connection between XPM and squeezing is not unknown [12]. A change in photon statistics, with the crucial use of a cavity, by using the Real part of XPM Kerr nonlinearity has been shown [14], [13]. In our system, it should be possible to see sub-Poissonian statistics of light, using the Imaginary part of the nonlinearity. This implies that this source of squeezing is novel. We show in fig. [11] that the enhancement of this source due to CPT (Coherent Population Trapping) makes it comparable to the linear absorption.

The question we have posed, and our calculations, lead us to postulate or conjecture three types of multiphoton processes:

Type1: When two or more fields face equal nonlinear susceptibilities, we show it could be the same multiphoton process that is visible in two or more fields simultaneously. It is counterintuitive because while the induced polarizations by each of the two fields can be unequal, the number of photons absorbed or emitted are equal when susceptibilities are equal (we show).

Type2: In the case of unequal susceptibilities, the multiphoton process should be visible only in one field. In the rest of the fields, because of equal number of absorption and simultaneous emissions of photons in the rest of the fields the process is not visible. We can show its suggested existence in system1.

Type3: the multiphoton process is visible only in one field due to the presence of a simultaneous reverse process that renders it invisible in other fields. We can show its suggested existence using system3 shown in fig[3].

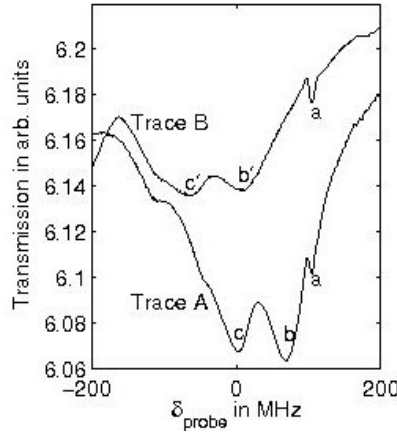


FIG. 4: This plot is from an experiment described in our previous work[18]. The narrow dip shown is newly interpreted as a nine-photon resonance. The other broad dips are linear absorptions. The typical intensity of probe and coupling lasers is about 2mWatt/.04 cmsq. and the weak counter signal beam is 40 μ Watt/.04 cmsq. It can be appreciated from this trace that the enhancement of this high order nonlinearity makes it conveniently observable. We expect it to be very sensitive to cross field correlations.

The system3 shows a $\chi^{(9)}$ nonlinearity in the beam providing the coupling Ω_{w24} . It is shown in the fig[4], obtained from an experiment done earlier[18] and was described as a three photon resonance but is newly interpreted as a nine photon resonance using this work. It is shown on the same trace that shows a linear feature. The strength of this high order nonlinearity is important because it is expected to be very sensitive to cross field correlations. The requirement is that there should be photons arriving simultaneously, that belong to different fields. When compared with SPM multiphoton process, this can change the nature of field statistics required for optimum results. XPM processes give a different information about the field statistics. A largely asymmetric nonlinearity can be used to experimentally test the conjectures of type2 and type3 multiphoton processes through cross field correlation sensitivity. It might tell whether virtual photons are in the picture instead.

II. THE SIMULATION

It is also important to note that the formalism to calculate susceptibilities that has been introduced by Harris et.al. often gives more than one solution. In such cases we either select the valid solution using physical arguments (for system2) or take the solution that matches with density matrix simulation[17] which we have for Schmidt et. al.'s system. The simulation is such that the frequency $\omega_b = \omega_c$. Therefore we compare the analytic results, for the special case when the Signal always has same frequency as the Coupling and the Coupling detuning changes, changing the position at which the Probe and the Coupling form EIT. We put the changing EIT positions or the coupling beam detuning on the X-axis. Both the analytical results and simulation are for zero velocity of atoms. The coupling detuning is from the upper excited level which is 121 MHz away from the lower excited state. The Probe detuning is from the lower state. For one case, the simulation is also shown with the ramping probe on the X-axis.

III. THE XPM NONLINEARITIES IN SYSTEM1

We follow Schmidt et.al.[8] and for steady state, after making the rotating wave approximation, write the following equations for system1:

$$-2\Delta\omega_{21}b_2 + \Omega_c b_3 + \Omega_{24}b_4 = 0 \quad (1)$$

$$\Omega_{31}^* b_1 + \Omega_c^* b_2 - 2Ab_3 = 0 \quad (2)$$

$$\Omega_{42}^* b_2 - 2Bb_4 = 0 \quad (3)$$

where $A=\Delta\omega_a-i\Gamma_3/2$ and $B=\Delta\omega_b-i\Gamma_4/2$ Here Ω 's are the Rabi frequencies and ω 's the frequencies of the laser beams and b 's are the probability amplitudes of various levels.

Case I:when all the atoms are in level $|1\rangle$ or EIT condition

For this case $b_1^*b_1=1$

1.nonlinearity faced by the probe

$$b_1^*b_3 = \frac{-4\Omega_{13}^*\Delta\omega_{21}A + \Omega_{13}^*|\Omega_{24}|^2}{-2|\Omega_{24}|^2A + 8\Delta\omega_{21}AB - 2|\Omega_c|^2B} \quad (4)$$

This is the only answer we get for this particular case. The polarization(ignoring the oscillations) $Nb_1^*\mu_{13}b_3$ at two photon detuning equal to zero is

$$\begin{aligned} & \varepsilon_0 \chi^{(5)} E_{13}E_{24}^4 + \varepsilon_0\chi^{(5)} E_{13}E_{24}^2E_c^2 \\ &= \varepsilon_0 \left[\frac{-2N\mu_{13}\mu_{13}^*|\mu_{24}|^4A^*}{\varepsilon_0\hbar^5 - 2|\Omega_{24}|^2A - 2|\Omega_c|^2B|^2} \right] E_{13}E_{24}^4 \\ &+ \varepsilon_0 \left[\frac{-2N\mu_{13}\mu_{13}^*|\mu_{24}|^2|\mu_c|^2B^*}{\varepsilon_0\hbar^5 - 2|\Omega_{24}|^2A - 2|\Omega_c|^2B|^2} \right] E_{13}E_{24}^2E_c^2 \end{aligned} \quad (5)$$

where μ 's are the dipole matrix elements, and N is the atomic number density and the terms in square brackets are the fifth order susceptibilities, $\chi^{(5)}$. Here, in eq(5) the second term gives the multi-photon process $\chi^{(5)}(-\omega_a, \omega_a, -\omega_b, \omega_b, \omega_c, -\omega_c)$, the emission $-\omega_a$, is out of the beam, thus making the absorption of a ω_a photon visible. μ_{ab} and μ_{ab}^* mean the absorption and emission of a photon respectively or vice-versa.

2.nonlinearity faced by the Coupling beam can be deduced from the following:

$$b_2^*b_3 = \frac{-\Omega_c^*|\Omega_{13}|^2|\Omega_{24}|^2B^*}{2BB^*DD^*} + \frac{\Delta\omega_{21}\Omega_c^*|\Omega_{13}|^2}{DD^*} \quad (6)$$

where

$$D = 4\Delta\omega_{21}A - \frac{|\Omega_{24}|^2A}{B} - |\Omega_c|^2 \quad (7)$$

This solution matches well with the simulation as shown in fig[5]and[6]

3.nonlinearity faced by the Signal beam can be deduced from the following:

$$b_2^*b_4 = \frac{|\Omega_c|^2|\Omega_{13}|^2\Omega_{24}^*B^*}{2BB^*DD^*} \quad (8)$$

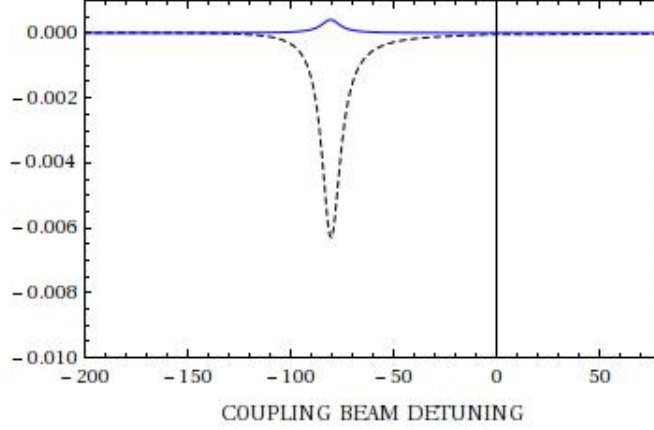


FIG. 5: This figure shows the numerically calculated absorption in solid line and dispersion in dashed line, in the coupling beam, in the Case I. The x-axis is the coupling (or signal beam detuning) beam detuning or the changing EIT position. The parameters are $\Omega_{13} = 0.1$ MHz, $\Omega_c = 3.55$ MHz and $\Omega_{24} = 5$ MHz.

The above solution matches well with simulation as shown in fig[7] and [8]

Case II: when probe and coupling have equal intensities or the CPT condition

For this case $b_1^* b_1 + b_2^* b_2 = 1$

We have put $\Delta\omega_{21} = 0$ in the following expression.

nonlinearity faced by the Signal beam can be deduced from the following:

$$b_4 b_2^* = \frac{\Omega_{24}^* - |\Omega_{24}|^2 A^* - |\Omega_c|^2 B^* B^*}{2(B^2 B^{*2} |\Omega_c|^2 |\Omega_{13}|^2 + B B^* - |\Omega_{24}|^2 A^* - |\Omega_c|^2 B^* B^*)} \quad (9)$$

We make a comparison with simulation when the rabi frequency of all the three fields is equal. We get the best agreement for the above solution, hence it is the valid one. The comparison with simulation is shown in fig.[9] and [10].

The nonlinearity here has an imaginary term that has $\Omega_{24}^* |\Omega_{24}|^4$ in the numerator. Squeezing of light will be hindered very little by the coexisting nonlinearity which has Ω_c in the numerator, because it is small and makes little difference.

Fig.[11] shows theoretical proof that this nonlinearity is comparable to linear features.

From the expressions above we see that different susceptibilities or combinations of $b_a b_b^*$ describe different multiphoton processes belonging to type 2, when the susceptibilities are unequal. This can be possible because the multiphoton process is visible in one of the beams of the system only, as it absorbs and emits a photon out of the beam, from the rest of the beams it absorbs and emits a photon, which replaces the absorbed photon exactly, which renders the process invisible. When the susceptibilities are equal, however, like in Case I, where the first terms in the expression for Coupling and Signal beams give

$$|Im[\chi^{(5)}]| = \left| \frac{|\mu_{23}|^2 |\mu_{13}|^2 |\mu_{24}|^2 \Gamma_4 / 2}{2 B B^* D D^*} \right| \quad (10)$$

(We have used the Absolute value since the complex conjugate of $\chi^{(5)}$ gives the opposite sign, the two signs refer to absorption and emission), then we show in a later Section, the symmetry suggests that the multiphoton process is type 1 rather than type 2.

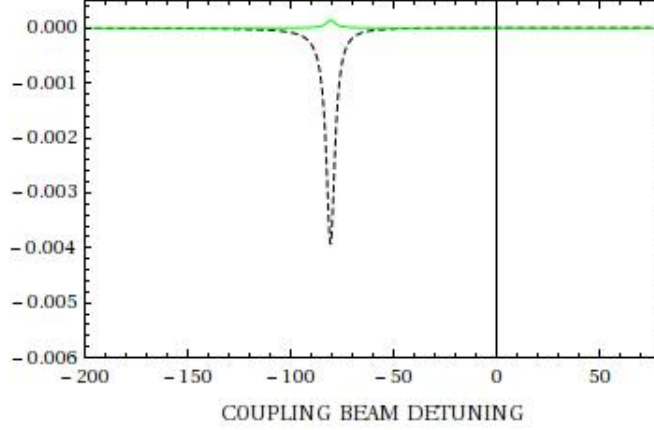


FIG. 6: This figure shows the analytically calculated absorption and dispersion in the coupling beam for Case I. The conditions remain the same as in fig[5]. The density matrix simulation treats mixed states while the analytical calculation pure states. The solutions match.

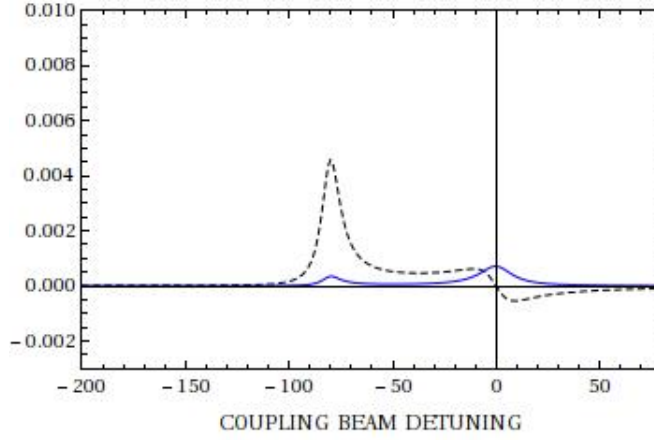


FIG. 7: This figure shows the numerically calculated absorption and dispersion in the Signal beam for Case I. The conditions remain the same as in fig[5]. The density matrix solution shows a linear absorption at detuning=0. For the analytic solution given in fig[8], $b_1 b_1^* = 1$, so there is no population in level $|2\rangle$, because of which there is no linear part. The rest of the solution matches.

IV. THE XPM NONLINEARITIES IN SYSTEM2

Following Harris et. al., we derive the following equations for system2, from the Schrodinger equation, after making the rotating wave approximation, in the steady state:

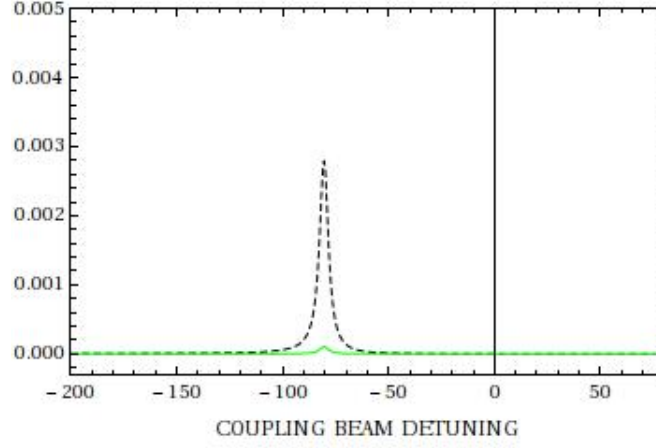


FIG. 8: This figure shows the analytically calculated absorption and dispersion in the signal beam for Case I. The conditions remain the same as in fig[5].

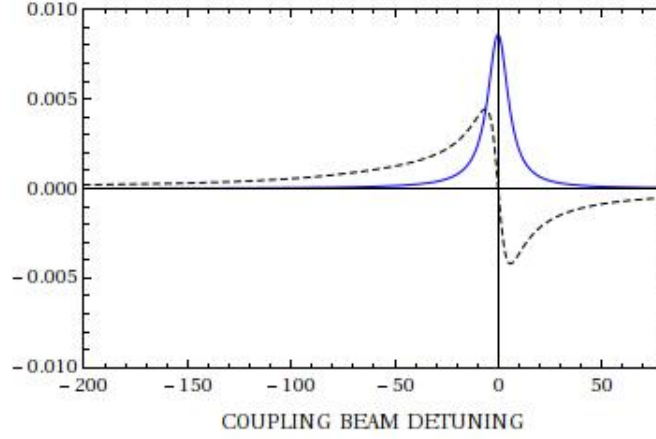


FIG. 9: This figure shows the numerically calculated absorption and dispersion in the Signal beam for Case II. The x-axis and the line types used remain the same as in fig[5]. The parameters are $\Omega_{13} = 0.1$ MHz, $\Omega_c = 0.1$ MHz and $\Omega_{24} = 0.1$ MHz. The analytic solution given in fig[10] fits it the best.

$$\Omega_{12}b_2 + \Omega_{13}b_3 = 0 \quad (11)$$

$$\Omega_{12}^*b_1 - 2\Delta\tilde{\omega}_{21} + \Omega_{23}b_3 = 0 \quad (12)$$

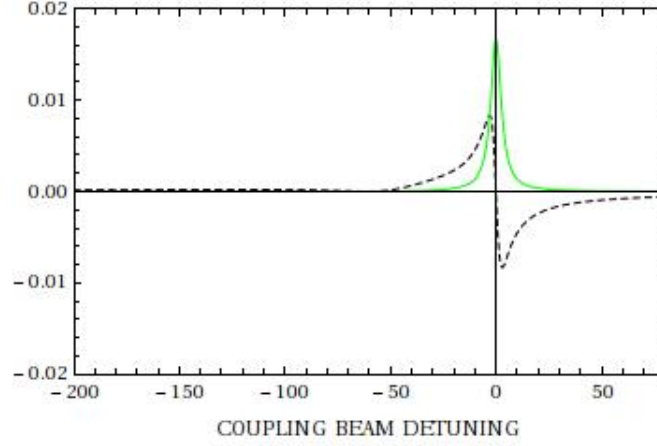


FIG. 10: This figure shows the analytically calculated absorption and dispersion in the signal beam for Case II. The conditions remain the same as in fig[9].

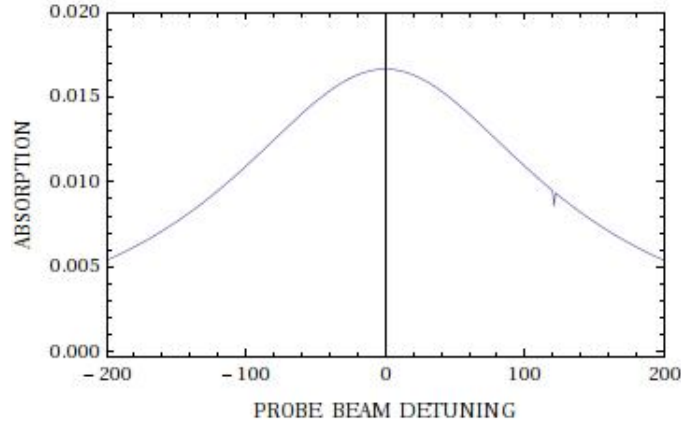


FIG. 11: This figure shows numerically calculated absorption in the signal beam for Case II. The parameters remain the same as in fig[9]. The coupling is in resonance with the upper excited state, 121 MHz away from the lower one. The atoms are in the uncoupled state so the linear absorption to an excited state is zero hence the lowest point of the CPT dip at probe detuning = 121 MHz shows the strength of the nonlinearity that causes squeezing. The analytic solution shows the coexisting nonlinearity makes very little difference. Hence this nonlinear absorption that can cause squeezing is seen to be comparable to the linear absorption.

$$\Omega_{13}^* b_1 - 2\Delta\tilde{\omega}_{31} + \Omega_{23}^* b_2 = 0 \quad (13)$$

where following Harris et. al. we define $\Omega_{12} = \Omega_{1i}\Omega_{i2}$, where i is the intermediate level, which is the rabi frequency corresponding to the fields ω_a and ω_b . Ω_{13} and Ω_{23} are the rabi frequencies corresponding to the fields ω_d and ω_c respectively. To identify the fields look at figure [2]. Also, $\Delta\tilde{\omega}_{21} = \omega_{21} - \omega_a - \omega_b - \iota\Gamma_2/2$ and $\Delta\tilde{\omega}_{31} = \omega_{31} - \omega_d - \iota\Gamma_3/2$.

nonlinearity faced by the field ω_d can be deduced from the following:

$$b_1^* b_3 = \frac{\Omega_{23}^* \Omega_{12}^* (4\Delta\tilde{\omega}_{31}\Delta\tilde{\omega}_{21} - |\Omega_{23}|^2)^*}{|4\Delta\tilde{\omega}_{31}\Delta\tilde{\omega}_{21} - |\Omega_{23}|^2|^2} + \frac{2\Delta\tilde{\omega}_{21}\Omega_{13}^*}{4\Delta\tilde{\omega}_{31}\Delta\tilde{\omega}_{21} - |\Omega_{23}|^2} \quad (14)$$

The first term is the non linear part. The occurrence of XPM fields in the form of single Ω 's in the numerator $\Omega_{23}^* \Omega_{12}^*$ indicates the absorption or emission of a single photon each from all the fields ω_c, ω_a and ω_b [9].

nonlinearity faced by the fields ω_a and ω_b can be deduced from the following:

$$b_1^* b_2 = \frac{\Omega_{13}^* \Omega_{32} (4\Delta\tilde{\omega}_{31}\Delta\tilde{\omega}_{21} - |\Omega_{23}|^2)^*}{|4\Delta\tilde{\omega}_{31}\Delta\tilde{\omega}_{21} - |\Omega_{23}|^2|^2} + \frac{2\Delta\tilde{\omega}_{31}\Omega_{12}^*}{4\Delta\tilde{\omega}_{31}\Delta\tilde{\omega}_{21} - |\Omega_{23}|^2} \quad (15)$$

The first term gives the nonlinear part.

nonlinearity faced by the field ω_c can be deduced from the following:

$$b_2^* b_3 = -\frac{\Omega_{12}^* \Omega_{32}^* \Omega_{13} (4\Delta\tilde{\omega}_{31}\Delta\tilde{\omega}_{21} - |\Omega_{23}|^2)}{\Omega_{23} |4\Delta\tilde{\omega}_{31}\Delta\tilde{\omega}_{21} - |\Omega_{23}|^2|^2} + \frac{2\Delta\tilde{\omega}_{21}}{\Omega_{23}} \frac{|\Omega_{23}^* \Omega_{13} + 2\Delta\tilde{\omega}_{31}^* \Omega_{12}|^2}{4\Delta\tilde{\omega}_{31}\Delta\tilde{\omega}_{21} - |\Omega_{23}|^2} \quad (16)$$

The first term gives the nonlinear part. To obtain the susceptibility $\chi^{(3)}, b_2^* b_3$ should be multiplied with μ_{23} and not μ_{23}^* . Only then the expressions in equations (14), (15) and (16) represent the possible multi-photon processes

$\chi^{(3)}(\omega_d, -\omega_a, -\omega_b, -\omega_c)$ or $\chi^{(3)}(-\omega_d, \omega_a, \omega_b, \omega_c)$, $\chi^{(3)}(\omega_c, \omega_a, \omega_b, -\omega_d)$ or $\chi^{(3)}(-\omega_c, -\omega_a, -\omega_b, \omega_d)$, $\chi^{(3)}(-\omega_a - \omega_b, \omega_d, -\omega_c)$ or $\chi^{(3)}(\omega_a + \omega_b, -\omega_d, \omega_c)$

The complex conjugates $b_3^* b_1, b_2^* b_1, b_3^* b_2$ describe the reverse multiphoton process.

We get from the first terms of the three equations:

$$\chi^{(3)} = \frac{\mu_{23}^* \mu_{12}^* \mu_{13} (4\Delta\tilde{\omega}_{31}\Delta\tilde{\omega}_{21} - |\Omega_{23}|^2)}{|4\Delta\tilde{\omega}_{31}\Delta\tilde{\omega}_{21} - |\Omega_{23}|^2|^2} \quad (17)$$

The absolute values of the first terms of $\text{Im}[\chi^{(3)}]$'s corresponding to the multi-photon processes $\chi^{(3)}(\omega_d, -\omega_a, -\omega_b, -\omega_c)$ or $\chi^{(3)}(-\omega_d, \omega_a, \omega_b, \omega_c)$, $\chi^{(3)}(\omega_c, \omega_a, \omega_b, -\omega_d)$ or $\chi^{(3)}(-\omega_c, -\omega_a, -\omega_b, \omega_d)$, $\chi^{(3)}(-\omega_a - \omega_b, \omega_d, -\omega_c)$ or $\chi^{(3)}(\omega_a + \omega_b, -\omega_d, \omega_c)$ are exactly equal. Though Harris et. al. have put $\chi^{(3)}(\omega_a, \omega_b, -\omega_d, \omega_c)$ and $\chi^{(3)}(\omega_b, \omega_a, -\omega_d, \omega_c)$ separately to be equal, we think that might not be true. We give our reasons later in this section. We also discuss how this case of "partial" symmetry is useful. The above nonlinearities, being equal, suggest type1 processes rather than type3. We show this in the next section. In type3 asymmetric processes the multiphoton process is visible in one beam only because there is a simultaneous reverse process that replaces the absorbed photons exactly with the emitted photons in the other beams. So the susceptibilities do not have to conform to any special symmetry condition, for the multiphoton process to belong to the type3. The same holds for type2. We note here that in type2 multiphoton processes it is not possible to have a reverse process that also has a emission out of the observed beam because, it is obvious, that changes the susceptibility of the system in a way that is not reflected in the calculations. (Then, the calculation shows only half the value of the actual susceptibility) So, the complex conjugate of susceptibility, for type2 processes, describes the same process instead of the reverse one.

The equality in Harris et.al.'s work[9] showing $\chi^{(3)}(\omega_a, \omega_b, -\omega_d, \omega_c) = \chi^{(3)}(\omega_b, \omega_a, -\omega_d, \omega_c) = \chi^{(3)}(\omega_d, -\omega_a, -\omega_b, -\omega_c)$, might be only partially true because while $\chi^{(3)}(\omega_a + \omega_b, -\omega_d, \omega_c)$ is symmetric with the other susceptibilities observed in ω_c and ω_d , the multiphoton processes separately seen in the fields ω_a and ω_b can have an asymmetric part apart from the symmetric part that should necessarily be there. This case is important as it shows how an asymmetric susceptibility can be seen as a sum of symmetric and asymmetric part.

V. GENERATION OF CROSS FIELD NOISE CORRELATION DUE TO THE SYMMETRY

$\mathbf{D} = \tilde{\mathbf{E}} + \varepsilon_0 \mathbf{P}$ where $\tilde{\mathbf{E}} = \mathbf{E}(\omega) e^{i\omega t}$

Symbols have the usual meaning.

Energy density U in electric media $= \frac{1}{2} (\mathbf{D} \cdot \tilde{\mathbf{E}}^*)$

$\partial U / \partial t$ in steady state is a constant and is equal to the energy taken away from the field by absorptions by the atoms. When there is no medium this quantity is zero, as the energy outflow due to propagating waves, estimated by the Poynting vector, is compensated by the energy inflow originating in the laser current.

ω_b in Schmidt et. al.'s system is one of the fields under observation and we look at the polarization it causes.

$\mathbf{P} = N \tilde{b}_4^* \mu_{24}^* \tilde{b}_2 e^{-i\omega_{0b}t}$ where N is the atomic number density and $\hbar\omega_{0b} = E_2 - E_4$ where E_2 and E_4 are energies of the levels marked as $|2\rangle$ and $|4\rangle$ in fig[1].

After making the rotating wave approximation, we have

$\mathbf{P} = N (b_4^* \mu_{24}^* b_2 e^{i\omega_{0b}t}) e^{-i\omega_{0b}t}$

Therefore, $\partial U / \partial t = n \hbar \omega_{0b} = \text{Re}[-\frac{1}{2} i \omega_{0b} N b_4^* \mu_{24}^* b_2 E(\omega_b)]$ where n is the number of photons absorbed or emitted and $\partial E / \partial t = 0$, in steady state.

Similarly, we find n for the other field ω_c . We show that the multiphoton process described by the equal susceptibilities given in eq(6) and eq(8) can be of type1 i.e., simultaneously visible in both the fields ω_c and ω_b , and a generation of cross field noise correlation, *For this we must show that the number of photons, n , absorbed or emitted from each field, is equal.*

$n \hbar = \text{Re}[-\frac{1}{2} i N b_2^* \mu_{23} b_3 E(\omega_c)]$, for field ω_c

(We actually mean only the first terms from the respective expressions of $b_2^* \mu_{23} b_3$ and $b_4^* \mu_{24}^* b_2$ since they only correspond to the multiphoton process we are considering)

We get

$$n = \left| - \frac{N |\Omega_c|^2 |\Omega_{13}|^2 |\Omega_{24}|^2 \Gamma_4 / 2}{4 B B^* D D^*} \right| \quad (18)$$

for both the fields, ω_c and ω_b . This can similarly be proved for all cases of the symmetry.

VI. XPM NONLINEARITY FOR THE CASE OF SYSTEM3

The consideration of this system leads to a postulation of existence of type3 multiphoton processes. The fig.[3] shows the relevant level scheme. After making the rotating wave approximation, we get the following equations:

$$-2\Delta\omega_{21}\Delta b_2 + \Omega_{23}\Delta b_3 + \Omega_{24}\Delta b_4 + \Omega_{w23}b_3 + \Omega_{w24}b_4 = 0 \quad (19)$$

$$\Omega_{31}^*\Delta b_1 + \Omega_{32}^*\Delta b_2 - 2A\Delta b_3 + \Omega_{w23}^*b_2 = 0 \quad (20)$$

$$\Omega_{42}^*\Delta b_2 - 2B\Delta b_4 + \Omega_{w24}^*b_2 = 0 \quad (21)$$

Since we use the results from system1 in solving for the nonlinearity, the rotating wave approximation should be the same as before. Δb 's are the perturbation probability amplitudes due to the weak couplings.

we solve the equations for the form $b_a^* \Delta b_b$ using the conditions

$$(b_1 + \Delta b_1)(b_1^* + \Delta b_1^*) + (b_2 + \Delta b_2)(b_2^* + \Delta b_2^*) = 1 \quad (22)$$

where $b_1 b_1^* = 1$ and

$$b_1^* \Delta b_1 + b_2^* \Delta b_2 = 0 \quad (23)$$

Solving the equations ,at two photon detuning zero, for $b_2^* \Delta b_4$,one of the solutions we get is :

$$\begin{aligned} [2|\Omega_{31}|^2|\Omega_{23}|^2 B + 2BDD^*]b_2^* \Delta b_4 \\ = + \frac{\Omega_{42}^* \Omega_{w23}^* \Omega_c |\Omega_c|^2 |\Omega_{13}|^2}{D} + \frac{\Omega_{w24}^* |\Omega_c|^4 |\Omega_{13}|^4}{|D|^2} \\ + \frac{2\Omega_{42}^* \Omega_{w24} \Omega_{24}^* |\Omega_c|^2 |\Omega_{13}|^2 A}{BD} - \frac{\Omega_{24}^* \Omega_c^* \Omega_{w23} |\Omega_{24}|^2 |\Omega_{13}|^2 A}{BD} \\ - \frac{\Omega_{w24}^* |\Omega_c|^4 \Omega_{13}^2}{D} \end{aligned} \quad (24)$$

For $b_4^* \Delta b_2$, one of the solutions we get is:

$$\begin{aligned} [2|\Omega_{31}|^2|\Omega_{23}|^2 BB^* + 2BB^* DD^*]b_4^* \Delta b_2 \\ = \frac{\Omega_{42} \Omega_{w24}^* \Omega_{24} A |\Omega_c|^2 |\Omega_{13}|^2}{D} + \frac{\Omega_{24} \Omega_{w23}^* \Omega_c B |\Omega_c|^2 |\Omega_{13}|^2}{D} \\ + \frac{2\Omega_{42} \Omega_{w23} \Omega_c^* A B |\Omega_c|^2 |\Omega_{13}|^2}{BD} \\ + \frac{\Omega_{24}^* \Omega_{24} \Omega_{w24} A B |\Omega_c|^2 |\Omega_{13}|^2}{BD} \end{aligned} \quad (25)$$

All solutions including the ones seen in Ω_{w23} field are similar to the extent that they show a ninth order nonlinearity and type3 multiphoton processes. We see from the simulation[18] done for system3 that an identical solution is not found in the Ω_{w23} field. Hence it is confirmed that the system3 shows multiphoton processes of type3 rather than type1. We note the occurrence of two Ω_{24} 's or Ω_{24}^* 's in the expression without its conjugate counterparts which would look like two consecutive photons getting absorbed(emitted) from the field. But it is accompanied with an emission(absorption) in the weak field, so it is not absorbing the second photon and going to a higher virtual level.

Since the nonlinearity absorbs photons from four different fields it is particularly sensitive to cross field correlations. Though the nonlinearities in system2 absorb photons from only three fields, they have the qualitative difference that they in no cases, at the same time require the absorption of more than one photon from a single field. These ninth order susceptibilities are shown by the simulation to be either largely asymmetric or completely asymmetric at selected detunings. So they can be used to test our conjecture of type2 and type3 processes against an alternative conjecture of virtual photons.

VII. PROPOSED EXPERIMENTS

An experiment to confirm the existence of multiphoton processes of type1 in the case of equal susceptibilities, can be done by mapping Schmidt et. al.'s system on to the hyperfine levels involved in the D2 line of Rb. The fields in which we expect the multiphoton process to be

visible simultaneously are the signal and coupling fields shown in fig.[1] The key to detecting type1 processes is the kind of fluctuation in the number of absorbing atoms that is analogous to shot noise in the case of radiation. Therefore, we must compare the fluctuations in the number of atoms that absorb radiation that is not related with the fluctuation in absorption due to shot noise, that will be the same in both fields only if the multiphoton process is type1. It should be noted that the fluctuations in absorption due to shot noise will be identical in the two fields due to the symmetry. We must emphasize the fluctuations in absorption will be completely identical only if the symmetry is a sufficient condition for type1 processes. For detection a concept inspired from relative intensity squeezing[2] might be useful. It is simple to confirm the existence of symmetries in the systems 1 and 2. we should look for cross field noise correlations. They exist whether or not the symmetry is a sufficient condition for the existence of multiphoton processes of type1. Very recent studies[3],[4] have shown cross field noise correlations in FWM situations.

Largely asymmetric nonlinearities can be used to test our conjecture of type2 and type3 process against a conjecture of virtual photons using the property of cross field correlation sensitivity.

VIII. CONCLUSIONS

To summarize, we postulate or conjecture three types of multiphoton process. While the type of multiphoton processes seen here are shown, to be either type2 or type3 (that are visible only in one field) when the susceptibility terms are asymmetric, when the susceptibilities faced by two fields show symmetry, then the multiphoton process might be type1 (that is visible in two or more fields simultaneously) rather than type2 or type3. Through these arguments we have seen a way to pick up the valid solution from the many solutions that Harris et. al.'s formalism often gives in system 2. Using our analysis here, we only partially agree with the first example of the symmetry, that Harris et. al.[9] give without going into the origin of this symmetry but find that the example is useful in deciding how an asymmetric case can be a sum of symmetric and asymmetric parts. We have shown that the symmetry generates cross field noise correlation. Type1 processes have the possibility of leading to strong sources of temporally entangled photons, of different frequency, as the nonlinearities are EIT enhanced. We have also shown, a $\chi^{(5)}$ XPM nonlinearity that can be a promising source of light squeezing as it is comparable to the linear absorption. It squeezes due to the Imaginary part of the susceptibility. We show this is unusual so this source is qualitatively novel. In the process of showing the suggestion of existence of type3 multiphoton processes, using a modified system, we show a way to generate an easily observable $\chi^{(9)}$ absorption resonance, which being a high order nonlinearity is expected to be very sensitive to cross field correlations. We have done an experiment described in our previous work[18] but the order of nonlinearity newly interpreted, that shows the $\chi^{(9)}$ feature, only an order smaller than linear absorption. We have shown how this nonlinearity can be used to test the conjecture of type2 and type3 processes.

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